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**ABSTRACTS OF RECENT PhD THESES  
FACULTY OF CHEMICAL ENGINEERING  
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**ABSTRACTS**

**FORMATION, DETECTION AND QUENCHING OF SINGLET OXYGEN**

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Electronically excited singlet molecular oxygen has a great importance in photochemistry and photobiology. There is strong evidence for the involvement of singlet oxygen, a powerful oxidant, in photosensitized oxidations, in phototherapy of cancer, in the photodegradation of dyes and polymers. My thesis consists of three subjects connected with the photochemistry of singlet oxygen.

The photophysical and photochemical properties of some new tetraphenylporphyrin type photosensitizers were determined in organic solution. The triplet quenching by oxygen is the most important step in the process of formation of singlet oxygen. The microenvironment of the sensitizer has an influence on the quenching process, for this reason quenching was studied not only in homogeneous solution but also in phospholipid bilayer. The bilayer of the vesicle is a simple, artificial model of the cell membrane. The quenching process is sensitive to the phase of the membrane and undergoes a significant change at the temperature corresponding to the main phase transition. The activation energies of the triplet quenching process were determined in the gel and fluid phase of the membrane. At a temperature below the phase transition the activation energy of the quenching process decreases as the polarity of the porphyrin increases. This observation is connected with the porphyrin localization in the lipid membrane because the viscosity change caused by raising the temperature is different in various parts of the membrane.

Nowadays singlet oxygen is detected almost exclusively by its weak near infrared (1270 nm) phosphorescence. A new measurement setup was built for the investigation of phosphorescence of singlet oxygen. It was possible to calibrate the near infrared emission signal to the concentration of singlet oxygen and to determine the phosphorescence radiative rate constant in different solvents. The

experimental data were evaluated by an own Monte Carlo simulation program and the light collection efficiency of our setup was investigated.

The quenching of singlet oxygen by coordinatively unsaturated (ligand)*bis*(1, 2-benzosemiquinonediiiminato)cobalt(III) complexes has been studied in different solvents. The investigation of the measured rate constants has proved that the quenching process is diffusion controlled. We applied the theory of the diffusion controlled reactions for the quenching process. The reactive distance of the encounter pair has been calculated to be 0.33 nm, shorter than the radius of the complex itself, which points to the fact that singlet oxygen must approach the central cobalt atom at the open axial position.

## **DETERMINATION OF KETONE BODIES AND CITRIC ACID IN COW'S MILK FOR DIAGNOSTIC PURPOSES**

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The energy deficiency in high-yielding dairy cows can cause subclinical or clinical ketosis, in which the concentration of ketone bodies (acetone, acetoacetate and  $\beta$ -hydroxybutyrate) increases in the different body fluids. The subclinical ketosis is associated with decreased body conditions and milk yield, impaired reproductive performance and increased risk of clinical ketosis. The early detection of elevated levels of ketone bodies during the early, subclinical stage of energy deficiency is highly recommended as on this basis appropriate corrections in the herd management can be introduced to reduce the development of clinical ketosis – consequently the profit-loss.

The aim of this study was:

- method development, optimisation and validation for reference and routine laboratory analysis of ketone bodies and citrate and
- clarification from diagnostic reason if the connection between elevated ketone body formation and citrate concentration in milk exists.

For these purposes:

- As reference methods new gas chromatographic (GC) and headspace gas chromatographic (HS-GC) methods were developed and validated for determination of acetone concentration in milk samples.
- The HS-GC method was developed with an appropriate three-steps sample-preparation for determination of acetate and  $\beta$ -hydroxybutyrate in milk.
- As a rapid method a flow injection system (FIA) was optimised and validated for determination of acetone in milk.
- Under real conditions of milk sampling, transport and storage, milk samples of 119 dairy cows were investigated. Based on the results of ketone bodies and citrate determinations the author established the following:
- the sample's acetoacetate content spontaneously decarboxylised to acetone during the traditional sample handling;
- the  $\beta$ -hydroxybutyrate and the acetone concentration showed an opposite change in the metabolically most critical first 6 weeks of lactation: the acetone content decreased, while that  $\beta$ -hydroxybutyrate increased in milk;
- significant positive correlation and linear connection between acetone and  $\beta$ -hydroxybutyrate levels were found in the samples with acetone concentration > 0.4 mmol/l;

- the author found a parallel drop of citrate and acetone content during the lactation, which can clearly be explained on the basis of biochemistry of tricarmonic acid cycle and ketogenesis;
- significant negative correlation between citrate and  $\beta$ -hydroxybutyrate and acetone content was also found in the samples with acetone concentration  $> 0.4$  mmol/l.

Focusing on these results, a routine laboratory analysis of milk citrate can be a useful tool in the early detection of energy deficiency in high-producing dairy cows.

## ECONOMIC AND CONTROLLABILITY ANALYSIS OF ENERGY-INTEGRATED DISTILLATION SCHEMES

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This work is based on studying and rigorous modelling (design, simulation, optimization, and control aspects) of various conventional and integrated distillation schemes for the separation of ternary mixture at different feed compositions. Economic evaluations of the distillation schemes are founded on rigorous simulation and optimization of each distillation scheme for minimum total annual cost (TAC). Thereafter, the controllability of the optimized schemes is studied, which includes the determination of steady state control indices, the open loop and closed-loop dynamic behavior. Overall evaluation of the best distillation schemes should be based on the combination of economic study, controllability study and environmental aspects. The main aims of this study are outlined below:

1. Design of various conventional and integrated distillation schemes, rigorous optimization of the investigated alternatives by steady-state simulation, comparison of the energy-integrated schemes and the best conventional distillation scheme for better energy utilization of TAC savings.
2. Design and simulation of Petlyuk column for different feed compositions and evaluation of its economic features for the given separation. A special emphasis was laid on the solution of the internal recycle streams, which represent an important design problem by pinpointing the theoretical fractional recovery ( $\beta^*$ ).
3. Design and simulation of the complex distillation arrangements (sloppy heat-integrated schemes), applying the theoretical fractional recovery ( $\beta^*$ ) concept for designing these complex distillation schemes.
4. Comparison of the effect of applying different price structures (European and U.S. prices) on TAC savings and ranking of the schemes.
5. Determination of the steady-state control indices of economically optimal distillation schemes at three different feed compositions and selection of the best control structure.
6. Investigation of the open loop and closed-loop dynamic behavior of the optimal distillation schemes at equimolar feed composition. Investigation of the closed-loop dynamic behavior for different control structures.
7. Estimation the flue gas emission reductions due to energy integration.

The Hyprotech's (HYSYS 1.1, 1996) professional simulation package is used for the modelling as well as steady-state and dynamic simulation of the distillation schemes. Steady-state control indices are determined by the use of MATLAB (Version 5.10.421, 1997).

The following methodological steps are implemented:

- a) Shortcut design procedures are used to estimate:
  - the number of theoretical trays,
  - feed location and draw-off trays,
  - and the reflux ratio.
- b) Steady-state rigorous simulation is carried out for the desired performance, the rigorous model of the package is also used for scaling the different equipment items.
- c) Steady-state control indices are determined for the economically optimal schemes at different control structures.
- d) Dynamic simulation is carried out for the optimized schemes under feed rate and composition disturbances.

The scope of this thesis work is to separate a multi-component mixture into pure components by means of continuous distillation. Design, simulation, operation, and control aspects of different distillation arrangements for the separation of ternary mixture are studied. Economic evaluations of distillation schemes are made by rigorous optimization of each distillation scheme for minimum total annual cost (TAC) using steady-state simulation. The controllability of the optimized schemes is studied, which includes the determination of steady-state control indices, the open loop and closed-loop dynamic behavior. Overall evaluation of the optimum distillation schemes are to be made based on the combination of economic study, controllability study and environmental considerations.

This study is devoted to a non-ideal alcohol system (ethanol, n-propanol, n-butanol) with high product purity (99%) and three different feed compositions (0.45/0.1/0.45, 0.33/0.33/0.33, and 0.1/0.8/0.1). The economic features of different distillation schemes are investigated including conventional distillation scheme, conventional heat-integrated scheme, Petlyuk column, and sloppy heat-integrated schemes and compared to the best conventional distillation scheme.

The investigated schemes are:

- i. Direct separation sequence without heat integration (**D**)
- ii. Direct separation sequence with forward heat integration (**DQF**)
- iii. Direct separation sequence with backward heat integration (**DQB**)
- iv. Indirect separation sequence without heat integration (**I**)
- v. Indirect separation sequence with forward heat integration (**IQF**)
- vi. Indirect separation sequence with backward heat integration (**IQB**)
- vii. Thermally coupled sloppy separation sequence (Petlyuk column) (**SP**)
- viii. Sloppy separation sequence with forward heat integration (**SQF**)
- ix. Sloppy separation sequence with backward heat integration (**SQB**)

The above mentioned schemes are designed and simulated rigorously, and then optimized for minimum total annual cost and compared with the best conventional distillation scheme for better energy utilization and savings in total annual cost (TAC). A particular emphasis is given to the question of fractional recovery

of the middle component in the prefractionator and as a consequence to the internal recycle streams in the complex distillation arrangements (Petlyuk column and sloppy heat-integrated schemes). The effect of using higher and lower utility prices (European and U.S. prices) on the energy and the total annual cost (TAC) savings are investigated for different types of energy-integrated schemes.

The controllability features of the distillation schemes that indicate good economic features are investigated by determining their steady state control indices. According to the calculated steady-state control indices, open loop and closed-loop dynamic simulations have been carried out for the equimolar feed to investigate its dynamic behavior. The time constants, controller settings, overshoots, and the settling times are determined to characterize the controllability features of the schemes.

Finally, flue gas emissions of the optimum schemes are estimated for two different types of fuels (natural gas and petroleum oil).

This work is a contribution to the evaluation of the various distillation arrangements for the separation of multicomponent mixtures considering their economic and controllability features. The work provides a better knowledge for the conventional and energy-integrated schemes. Energy requirements and controllability are the most emphasized design aspects. The complex distillation arrangements (Petlyuk column and sloppy heat-integrated schemes) are very attractive from the energy point of view but their complexity makes them rarely used processes. The study of their properties has become principal parts of this work. This study can be considered as an extension to the previous studies in the area of energy integration and controllability of distillation columns.

## MODELLING PHASE EQUILIBRIA BY EQUATIONS OF STATE

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For designing separation processes the chemical engineers should be able to describe phase equilibria. I tried to develop equations of state (EOS) models based on molecular thermodynamics, so they are hopefully suitable for extrapolation of thermodynamic data.

The models consider:

- size and shape of molecules that are built of groups, giving a repulsion term,
- non-specific (van der Waals) attraction between groups,
- specific attraction between groups.

Combining the above mentioned parts I developed four EOS variants:

- Carnahan–Starling+van der Waals + Smirnova–Victorov (CS–vdW–SV),
- Boublík–Nezbeda+van der Waals + Smirnova–Victorov (BN–vdW–SV),
- Boublík-II. + van der Waals+Smirnova–Victorov (B-II.–vdW–SV),
- Boublík + Alder–Chen–Kreglewski+Smirnova–Victorov (BACK–SV).

I compared the EOS variants for pure component vapour pressure and equilibrium density data of n-alkanes, alpha-ethers and n-alkanols. With the best two EOS variants I predicted mixture phase equilibria data both at low and elevated pressures.

New scientific results:

1. I developed a group contribution method for building molecular parameters of the four EOSs mentioned, using the original Boublík model for alpha non-sphericity parameter.
2. Comparing the different group contribution model versions I found that the BN–vdW–SV and the BACK–SV EOSs are the best for pure components.
3. I developed a new group contribution model for the alpha non-spherical parameter of Boublík repulsion term. I evaluated the values of parameters of new models for CH<sub>3</sub>, CH<sub>2</sub> and OH groups from experimental data.
4. Using the parameters calculated from pure component data I predicted phase equilibrium data of mixtures. I compared different mixing rules for repulsive parameters of the two best models, and selected the best mixing rules. I investigated the possible reasons of faults of extrapolation, and made suggestions for further development.



## MODELLING OF THE STRUCTURE OF ARSENIC AND GERMANIUM CHALCOGENIDE GLASSES AND INVESTIGATION OF THEIR APPLICABILITY IN FIBER OPTICS

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The aim of our investigations was the examination of the following class of chalcogenide glasses: arsenic and germanium sulfides as well as their selenides and tellurides. These materials offer a wide range of industrial applications. Therefore, it was worth to extend our knowledge on this class of glasses.

The literature refers to the general structural characteristics of the chalcogenide glasses. According to these data the structure of these glasses lies between those of crystalline and amorphous materials. In the latter only a short range order has been observed. In contrast to this, our experience suggests that certain correlation exists also beyond the first coordination sphere suggesting a middle range order and the cluster model. According to this model these glasses are composed of 15–20 Å sized structural units, called ‘clusters’, which consist of ordered short range units. Our investigations were based on this concept and some other literature data.

In the case of arsenic sulfide, selenide and telluride these units are the  $\text{AsX}_3$  pyramids, while the structures of the germanium sulfide, selenide and telluride consist of tetrahedral ( $\text{GeX}_4$ ) units.

Our further examination was based on this observation of general literature data.

During the course of our studies the purpose was the quantum chemical modelling of different possible cluster types and to calculate their vibrational spectra, then comparing the resulted data with the experimental ones and to reveal the possible real structural units among the clusters of the glass in question.

The following possible cluster types were found for arsenic chalcogenides:  $\text{As}_2\text{X}_5$ ,  $\text{As}_4\text{X}_6$ ,  $\text{As}_6\text{X}_9$  [ $\text{X} = \text{S}, \text{Se}, \text{Te}$ ]. In the case of germanium chalcogenides the corresponding units were the  $\text{Ge}_3\text{X}_6$ , the  $\text{Ge}_4\text{X}_8$ , the  $\text{Ge}_5\text{X}_{10}$  and the  $\text{Ge}_6\text{X}_{12}$  clusters.

We elaborated the results in two steps: for a better assignment of the vibrational frequencies we carried out a normal coordinate analysis, then simulated the infrared spectra with the help of the quantum chemically calculated IR intensities. In this way we could determine the frequencies and characters of the individual vibrational modes (i.e. the weights of the internal coordinate types in it) and the intense absorption bands of the glasses. These could give important information for their further application.

The results of these calculations were that the structures of the clusters of the same size are very close: the valence and torsional angles are nearly identical in the

corresponding arsenic and germanium selenide, sulfide and telluride clusters. With increasing masses of cluster's atoms the frequencies of the individual vibrational modes shift to the lower region. In the same way with increasing substituent atom masses the characters of vibrational modes were changed, the participation of one of the internal coordinates types increased in the given mode.

We compared the simulated IR spectra of the investigated glasses in different aspects. We established a method to compare one another since their absorption ranges are different. In this way we offer information about their possible applications.

We examined some three-component materials preferred by the fiber optics. Significant differences can occur between the isomers depending on the positions of the different atoms. The complex glasses are not simple physical mixtures: we compared the mixed IR spectra of two glasses with the spectra of corresponding 'chemically mixtured' cluster.

We hope that our results will help in the more extensive application of the investigated glasses since the optical fibres participate more and more in our everyday life.

## OCTANE REFORMING OVER CATALYSTS MODIFIED BY ADSORBED METALS

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The PhD thesis was made at the Department of Chemical Technology of the Budapest University of Technology and Economics in close co-operation with MOL Plc. An industrially used reforming catalyst (Pt-Sn) was modified by adsorbed metals (palladium, gold, iridium, tin, tellurium and bismuth) and tested under close to industrial conditions. This modification-technology, the metal adsorption, was not used formerly in industrial research. The advantage of the technology is the close contact between the platinum and the modifying metal, since the modifying metal is adsorbed directly onto the platinum surface.

During the characterisation of the catalysts it was revealed that after the modification the adsorption took place differently from that mentioned in the literature. We cannot wash out all ions from the catalyst, some solution remains in the pores. During the drying step these ions impregnated the surface of the support. The hydrogen spill-over also has its influence on the 'over-modification'. The added metal above the theoretical amount and the metal placed selectively onto the platinum surface by adsorption both had their own effect on the catalyst properties.

The impact of the tellurium and bismuth was a decreased catalytic activity. Positive effect was exerted, however, on the selectivity, since the aromatic content of the liquid product decreased significantly, while that of the isoparaffins increased. From the industrial point of view these catalysts have low activity and their liquid yield is small. Their working mode is the adsorption on the platinum surface, so they selectively block the metallic sites. Reactions taking place on the acid sites of the catalyst become dominant among the possible reaction pathways (hydrocracking, isomerisation).

As the palladium, gold, iridium and tin modified catalysts had similar activities as the reference catalyst, they were characterised by increased space velocity. The iridium and gold modified catalysts were more active than the reference one. After the comparison of the liquid and gas yields at different temperatures, it turned out that the hydrocracking selectivity of the modified catalysts is only slightly dependent on the temperature, while the reference catalyst produced significantly less liquid product at higher temperature. The inactive gold splits the platinum surface into smaller clusters, so the selectivity is pushed toward the non-destructive reactions. Both metals increase the aromatic and decrease the isoparaffin content of the reformat. This result is the opposite of the basic objectives.

The other two modifiers, tin and palladium, have a positive impact on the catalyst selectivity. As the result of adsorbed tin liquid yield was higher at higher temperature, than that with the reference catalyst. At lower temperature the aromatic content of the reformat decreased, and the isoparaffin content increased. The

interpretation of this is the hindered metallic function that is supported by the amount of the naphthenic hydrocarbons (twice as the reference). The palladium modified catalyst produced a similar amount of liquid product as the reference one. However, the isoparaffin/aromatic ratio increased at higher temperature, while at lower temperature it remained practically unchanged. At higher temperature the advantageous selectivity change is more significant from the industrial point of view. The activity of the palladium and gold modified catalysts decreased during some oxidative regeneration cycles.

## DIASTEREOSELECTIVE HETEROGENEOUS CATALYTIC HYDROGENATIONS

Viktor HÁDA

Supervisor: Antal Tungler

*Department of Chemical Technology*

My PhD work was started as a new research project, the diastereoselective heterogeneous catalytic hydrogenation of *N*-heterocyclic aromatic compounds. Among the nitrogen containing heterocyclic compounds there are several optically active derivatives, which are important and valuable pharmaceutical intermediates or products. For example with the heterogeneous catalytic hydrogenation of picolinic acid derivatives the precursors of local anaestheticum molecules (Mepivacain, Bupivacain) can be obtained. Usually, after the heterogeneous catalytic hydrogenation the optically active product is prepared by optical resolution, in these cases the asymmetric hydrogenation is offered alternatively.

First, the substrate compounds were synthesized. In the hydrogenations the effect of different reaction parameters, such as active metal, solvent, temperature and pressure was studied on the diastereoselectivity. The qualitative and quantitative analysis of the hydrogenation products was performed by NMR, CD and HPLC measurements.

As pyridylglycols possess pharmacological effect, the heterogeneous catalytic hydrogenation of 2,2'-pyridoin was also studied in detail.

In both the hydrogenation of *N*-heterocyclic aromatic rings and the group bridging of the aromatic rings of the 2,2'-pyridoin the main question was how the built-in and available chirality elements influence the stereoselectivity of the reaction, namely the diastereoselectivity. The other important question was, which methods are available for the researcher to achieve the best stereoselectivity, arising from the selection of catalysts, the preparing of the catalysts and the selection of the reaction conditions.

In the hydrogenation of 2,2'-pyridoin and its derivatives there was larger 'free-space' to influence the stereoselectivity of the reaction by 'catalytic' means, since the active metal, the catalyst support and also the solvents had an effect on the diastereoselectivity. In fact, in the saturation of aromatic rings only 'coarse' control is possible, in the hydrogenation of 2,2'-pyridoin 'fine' tuning is also possible. From this point of view it is similar to the enantioselective hydrogenations carried out over modified metal catalysts.

The heterogeneous catalytic hydrogenation of chiral pyridine and pyrrole derivatives could be carried out in non-acidic medium. The (*S*)-proline-based substituent proved to be an effective synthon inducing high diastereoselectivity. In the diastereoselective hydrogenations mainly the structure of the substrate molecule determines the diastereoselectivity. Considering both the activity of the metal catalyst and the diastereoselectivity of the hydrogenation, palladium is the best catalyst

for the saturation of pyridine ring, while rhodium is the best one for that of pyrrole ring. The secondary and tertiary amines – the products of the hydrogenation of pyridine and pyrrole compounds – act as catalyst modifiers, increasing the *d.e.* at higher conversion values, in the last period of the reaction the hydrogenation is completely diastereoselective. Due to the less rigid molecular structure of the pyrrole derivative, compared with the structure of pyridine compounds, in the hydrogenation of the pyrrole compound the temperature had a significant effect on the diastereoselectivity.

In the hydrogenation of 2,2'-pyridoin with palladium on carbon the main hydrogenation product is the *meso*-hydroypyridoin because of extensive isomerization and the difference in hydrogenation rates for the *trans*- and *cis*-enediols. Platinum, rhodium and ruthenium possess much lower isomerization activity, consequently in the hydrogenations – via the *trans*-enediol – the *dl*-hydroypyridoin was formed, in excess. In the hydrogenation of 2,2'-pyridoin, the best catalyst for producing *meso*-hydroypyridoin was palladium black and that for the *dl*-isomer was rhodium on carbon. Supported platinum catalysts gave different *d.e.* values for both the *meso*- and *dl*-hydroypyridoins, which was explained by the competing enolisation and C = O reduction. The hydrogenation of 2,2'-pyridil occurs via 2,2'-pyridoin intermediate. We suppose the same hydrogenation mechanism on the catalyst surface because of the same chemo- and diastereoselectivity. In the hydrogenations of aromatic *N*-heterocycles the applied solvents influenced the activity of metal catalysts, whilst in the hydrogenation of 2,2'-pyridoin the solvents had also a significant effect on the diastereoselectivity.

During evaluation of the results the question arises whether the results of the experiments can be made use of in the practice, in order to prepare optically pure derivatives. The reactions carried out with complete diastereoselectivity can be applied in the practice, if the product and the chiral synthon can be separated with hydrolysis and with the following separation methods. Another question is whether racemisation occurs during these procedures. Concerning the products obtained from the reactions with non-complete stereoselectivity, the optical resolution comes up, since it is well-known that the enantiomers can be separated with better results from the mixtures containing them in a ratio different from 1:1 than from the racemic mixture.

## DETERMINATION OF URANIUM AND TRANSURANIUM ELEMENTS IN RADIOACTIVE WASTES OF LOW AND MEDIUM ACTIVITY FROM A NUCLEAR POWER PLANT

Sándor TÖLGYESI

Supervisor: Iván Gresits

*Department of Chemical Technology*

I developed a new analytical procedure for the alpha-spectrometric determination of uranium ( $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{234}\text{U}$ ) and transuranium isotopes ( $^{239+240}\text{Pu}$ ,  $^{244}\text{Cm}$ ) in low- and medium-activity liquid radioactive wastes, including exhausted ion-exchange resins, sludges and evaporation residues formed during the operation of the VVER-440 reactor of the nuclear power plant in Paks, and in waters to be released into the environment, which should be, according to the technology, inactive.

The main steps of the procedure developed are as follows:

- sample preparation based on chemical separation,
- production of a thin-layer prepareate,
- analysis by alpha spectrometry.

Analysis of  $\alpha$ -emitting isotopes in radioactive wastes from nuclear power plants is technically not a simple task. In some cases due to the low radioactive concentration (in waste waters for  $^{238}\text{U}$  0.5–1.0  $\mu\text{g/l}$ ), or to the difficult matrix environment (for ion-exchange resins and sludges) highly efficient enrichment or complicated chemical separation become inevitable. Hence, the heel of Achilles of the analytical procedure here is sample preparation.

The other crucial point of transuranium analysis is the production of a thin-layer prepareate for the energy-selective  $\alpha$ -spectrometric measurement. The small sample obtained after enrichment is to be spread on a highly polished stainless steel disc in a well-adhering form as a layer of uniform thickness which is infinitely small with respect to the transmission of  $\alpha$ -rays, in a way that the radionuclide to be measured is situated at the surface of the prepareate.

The results of the work aimed at the development of new methods for the sample preparation to the alpha-spectrometric analysis of liquid radioactive wastes from nuclear power plants can be summarised as follows:

- Ion-exchange resins were decomposed in oxygen atmosphere at overpressure in a closed calorimeter bomb. I have found no paper in the literature using such technique for the sample preparation of radioactive wastes.
- The solid fraction of radioactive sludge samples was treated, following decomposition, by extraction and re-extraction, using an extractant with apolar solvent base and containing tributyl phosphate. Although tributyl phosphate is widely applied as an extractant, this type of procedure was first developed and applied by myself as a reactor-specific method to the determination of uranium and plutonium in low- and medium-activity radioactive liquid waste (sludge) samples from a nuclear power plant.

- I worked out a new selective method for the determination of the alpha-emitting constituents in evaporation residues containing both organic and inorganic compounds and saturated with boric acid.
- The organic compounds present in radioactive sludges, evaporation residues and waters to be released into the environment were decomposed in an Erdey-Jankovits digester modified by myself for my specific purposes. The modified digester, in which the entire sample preparation procedure involving chemical separation can be carried out, is universally applicable for sample preparation for the determination of alpha-emitting isotopes in any type of low- and medium-activity liquid radioactive waste occurring in a nuclear power plant.
- I developed and was the first to apply an electrodeposition technique for the production of thin-layer prepares to alpha-spectrometric analysis of radioactive waste streams in the nuclear power plant in Paks. Uranium and transuranium isotopes were deposited by electrolysis in a device designed and constructed in our laboratory with a thermostat and controlled cathode potential adjusted at different values for each element to be deposited. The accuracy of the results of determinations carried out by means of the sample preparation and analytical methods developed was tested in the framework of the international QUALITY ASSESSMENT PROGRAM (QAP) organized by the U.S. Department of Energy Environmental Measurements Laboratory (U.S. DOE EML) for evaluating the results of environmental radiological measurements.
- The test results show that the accuracy of the results of measurements made by means of the methods developed meet the requirements set by QAP.
- The test results prove that the methods are reliable and can be applied for the routine determination of the activity concentrations of alpha-emitting isotopes in low- and medium-activity radioactive wastes formed in the VVER-440 type reactor and in waste waters released into the environment.
- Using the new analytical methods developed, we regularly measure the uranium and transuranium radionuclides in the radioactive liquid wastes, released water, and in the water in test wells bored around the nuclear power plant in Paks.